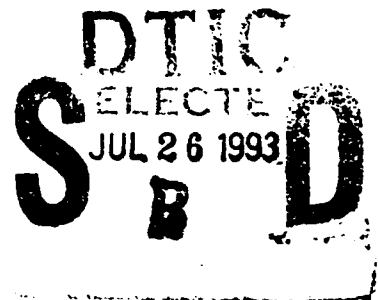AEOSR·TR· ن 3   ٥549.

MERGED BEAM STUDIES OF LASER STIMULATED

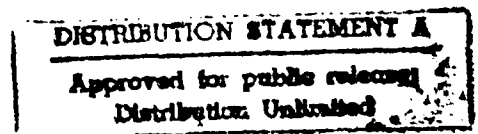# RADIATIVE RECOMBINATION

4th Technical Report

30 June 1993

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REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION	1b. RESTRICTIVE MARKINGS
Unclassified	

2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION / AVAILABILITY OF REPORT
2b. DECLASSIFICATION / DOWNGRADING SCHEDULE	unlimited

4. PERFORMING ORGANIZATION REPORT NUMBER(S)	5. MONITORING ORGANIZATION REPORT NUMBER(S)
	AFOSR

6a. NAME OF PERFORMING ORGANIZATION	6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION
The University of Western Ontario		Same as 8a

6c. ADDRESS (City, State, and ZIP Code)	7b. ADDRESS (City, State, and ZIP Code)
Dept. of Physics London, Ontario, Canada N6A 3K7	Same as 8c

8a. NAME OF FUNDING / SPONSORING ORGANIZATION	8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER
AFOSR	NE	F49620-92-J-0251

8c. ADDRESS (City, State, and ZIP Code)	10. SOURCE OF FUNDING NUMBERS			
	PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT ACCESSION NO
Bolling AFB DC20332-6448	61103F	2301	DS	

11. TITLE (Include Security Classification)

Merged Beam Studies of Laser Stimulated Radiative Recombination

12. PERSONAL AUTHOR(S) J.B. Mitchell

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Year. Month, Day)	15. PAGE COUNT
Annual	FROM 6/92 - 5/93	30 June 1993	

16. SUPPLEMENTARY NOTATION

17.	COSATI CODES		18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB-GROUP	

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

New measurements of the stimulated recombinations of electrons and protons to form hydrogen atoms in states n=11, 12 and 13 are presented. Measurements have also been made for the stimulated recombination of electrons and He^+ to form n=15 and n=16 states. These cross sections show structure due to singlet and triplet capture. The effects of laser radiation on the dissociative recombination of molecular ions H_3^+ have been examined and the results are reported.

20. DISTRIBUTION / AVAILABILITY OF ABSTRACT	21. ABSTRACT SECURITY CLASSIFICATION
☐ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☒ DTIC USERS	Unclassified

22a. NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE (Include Area Code)	22c. OFFICE SYMBOL
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DD FORM 1473, 84 MAR   83 APR edition may be used until exhausted.
All other editions are obsolete.

INTRODUCTION

Over the past three years, the Merged Beams Laboratory at the University of Western Ontario has been engaged in a project to demonstrate and to characterize the laser induced enhancement of the rate for the radiative recombination of electrons and protons. The primary impetus for this study is to develop a method for producing neutral antihydrogen atoms by recombining positrons and antiprotons in an efficient way. The work performed to date, and for the foreseeable future however, has dealt only with normal matter, i.e. electrons and protons. The ability to alter the rate of a chemical reaction by shining laser light onto the reagents is of fundamental interest in its own right. Thus, in recent studies, we have extended our original mandate to include studies of laser stimulated radiative recombination of other atomic ions such as He^+. We have also begun to examine the effects of photon illumination of molecular ions undergoing recombination with electrons. Here we have found a new effect in which the illumination leads to a reduction in the recombination rate. This is a very fertile area of investigation and will undoubtedly have numerous applications.

LASER STIMULATED RADIATIVE RECOMBINATION

When an electron makes a collision with an atomic ion, one can picture the electron as existing temporarily in an extended Bohr orbit during its brief encounter with the ion core. This can be represented as

$$e + A^+ \rightarrow A(nl) + h\nu,... \quad..........(I)$$
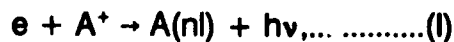
where n and l are the principal and orbital angular momentum quantum numbers respectively. For a thermal electron, the time of interaction is very short (10^{-15} secs). If during this time period, the electron drops down to a lower Bohr orbit, a photon will be emitted and if this lower orbit is bound, the recombination will be stabilized. Photon emission times are typically of the order of 10^{-9} seconds however, so this event is very rare. This means that radiative recombination occurs with a very small reaction rate.

The cross section for radiative recombination to a particular n state can be calculated using Kramers formula[1]:

$$\sigma = 2.1 \times 10^{-22} cm^2 \frac{E_0^2}{nE_{CM}(E_0 + n^2 E_{CM})}, \ldots (1)$$

where E_0 is the binding energy of the ground state of the ion, i.e for H this is 13.6 eV. E_{CM} is the center-of-mass collision energy.

If the electron-ion collision takes place in the presence of a photon field, it is possible for the photons to pump the stabilizing transition to a lower Bohr orbit if their energy is equal to the energy separation between the upper and lower states. This can lead to a greatly enhanced recombination rate. This process is known as *Laser Stimulated Radiative* Recombination i.e.

$$h\nu + e + H^+ \rightarrow H(nl) + 2h\nu, \ldots\ldots\ldots\ldots(II)$$
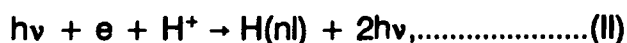
The degree of enhancement produced by the photon illumination can be estimated from the ratio of the Einstein B and A coefficients for stimulated and spontaneous transitions respectively[2]. Thus for laser power P and beam cross sectional area F, the gain factor, (the ratio of the cross sections for stimulated to spontaneous recombination

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into a specific nl level) is given by:

$$G = \frac{Pc^2}{8\pi F h \nu^3 \Delta \nu}, \ldots (2)$$

where ν is the frequency of the stimulating and emitted photon and $\Delta \nu$ is a frequency or energy spread term. The smaller $\Delta \nu$, the better the degree of matching between the pumping frequency and the energy separation between the initial and final states. In practice the value of $\Delta \nu$ is limited by (a) the frequency width of the laser beam, (b) the energy width of the states and (c) the spread of the collision energy in the center-of-mass reference frame. In practice, (c) usually dominates.

$\Delta \nu$ is related to ΔE_{CM} by the relation

$$\Delta \nu = (m v_e / h) \Delta v_e = \Delta E_{cm} / h, \ldots (3)$$

Figure 1 illustrates the radiative recombination process in terms of an energy level diagram. It can be seen that for a fixed laser frequency, stimulation to a given atomic energy level, n_a, will occur for a fixed collision energy, E_a. If the collision energy is increased to E_b where:

$$E_b - E_a = E_0 \left(\frac{1}{n_b^2} - \frac{1}{n_a^2} \right), \ldots (4)$$

stimulation will occur to the next higher state, n_b. E_a is given by:

$$E_a = h\nu - E_0 / n^2, \ldots (5)$$

Stimulated Radiative Recombination
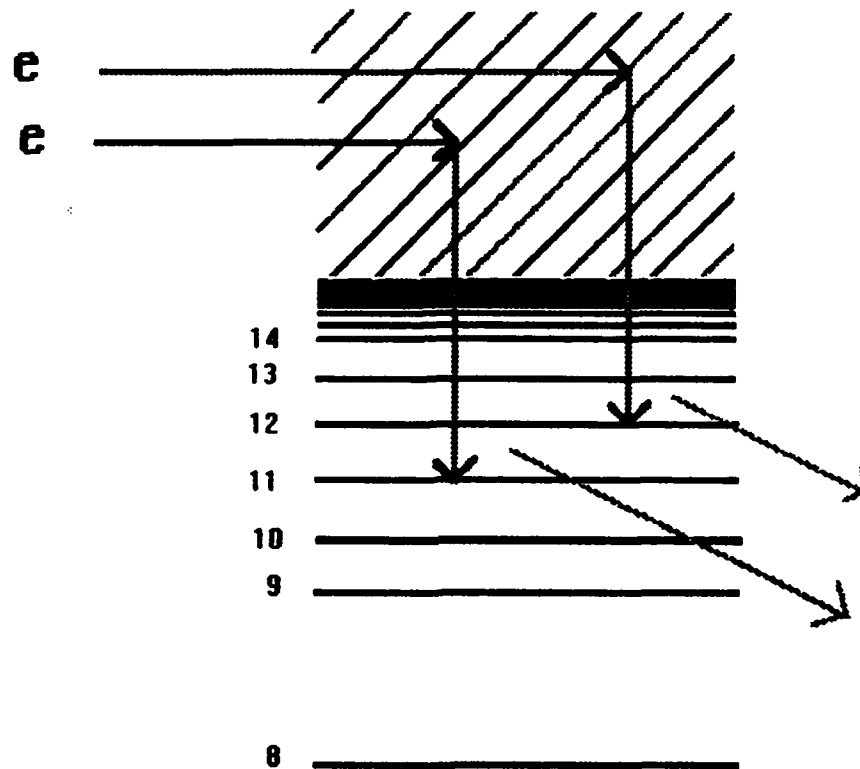
1. Energy level diagram representation of the stimulated radiative recombination process.

PREVIOUS WORK

The first experimental demonstration of *Laser Stimulated Radiative Recombination* (LSRR) was reported by us in 1991[3]. In that experiment, which employed a 10.6 μm carbon dioxide laser operating with the Merged Electron-Ion Beam Experiment at the University of Western Ontario, (Figure 2), stimulated recombination of electrons and protons to form hydrogen atoms in principal quantum number states n=11 and 12 was measured. In a similar experiment employing a pulsed dye laser and an ion storage ring at the University of Heidelberg, Schramm et al.[4] measured the stimulated radiative recombination to form H(n=2).

The original experiment at UWO involved a single passage of the laser beam across only about 1/80th of the electron-ion interaction region. While this was sufficient to demonstrate the efficacy of the process, the resulting signal was small necessitating long counting times to achieve satisfactory statistical accuracy. In an effort to improve this situation, the multipass cavity shown in figure 3 was constructed and installed in the interaction region. This device as tested, allows the laser beam to make a total of fifteen passes across the interaction region thus increasing the distance over which the electrons, ions and photons interact, and therefore the signal detected, by a corresponding amount.
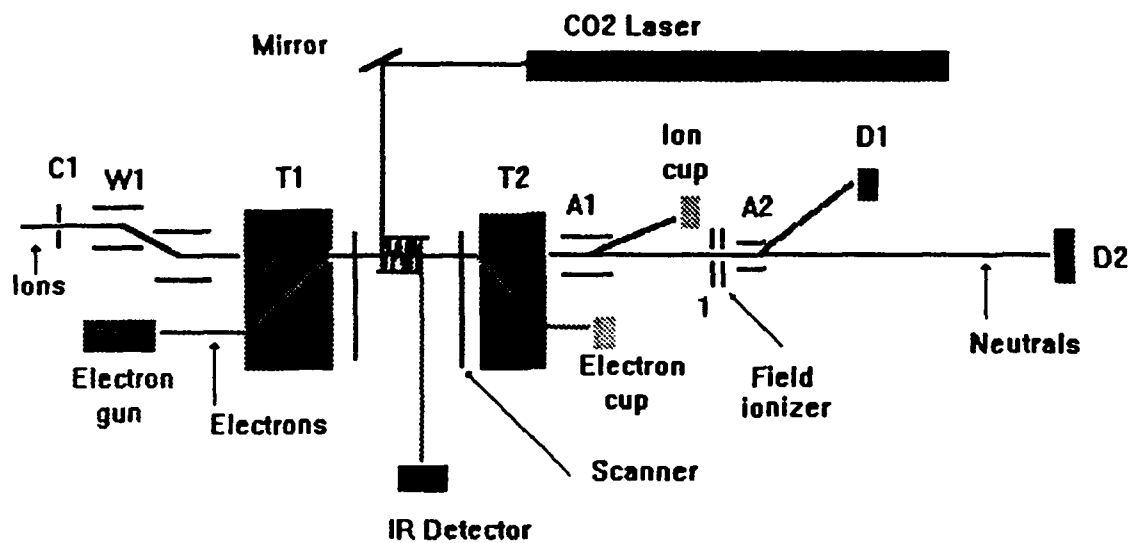
2.   Schematic diagram of the Merged Electron Ion Beam Experiment (MEIBE II) at the University of Western Ontario.

Photons   e + Ions

I.R. Detector
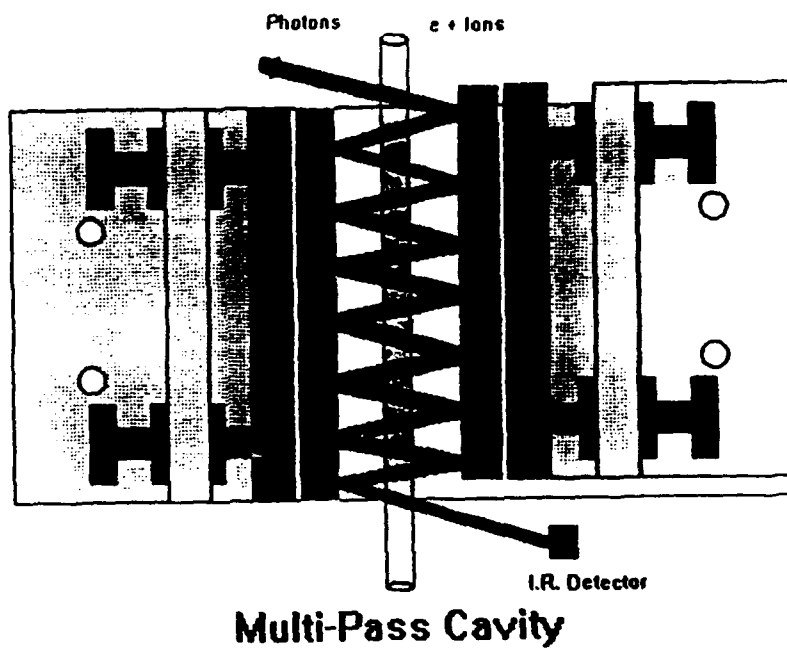
Multi-Pass Cavity

3.   Multi-pass cavity used to increase the illumination of the interaction region in the MEIBE II apparatus.

CURRENT STUDY

The current report describes new measurements of the stimulated recombination of electrons and protons to form hydrogen atoms in states with n=11, 12 and 13. We have also investigated the relationship between gain and laser power over the range from 0 to 15 watts.

As mentioned above, we have extended our studies to other ions and report here the first measurement of the stimulated recombination of electrons and He^+ ions. A new feature that is seen in these results is the recombination to form atoms in different spin states.

Lastly we have examined the effect of laser illumination on the rate of dissociative recombination processes using H_3^+ as a test ion.

RESULTS AND DISCUSSION

1. Electron-Proton Stimulated Recombination

The new multipass cavity, (Figure 3), installed in the MEIBE II apparatus, (figure 2), has been used to enhance the illumination of the interaction region by a factor of fifteen. This in turn has resulted in a signal enhancement of a factor of fifteen, allowing for much faster data collection and corresponding ease of operation. Using this device, we have remeasured the stimulated recombination of electrons and protons to form hydrogen atoms in the n=11 and 12 states. We have further extended the measurements to include recombination to the n=13 state. The results are shown in figure 4. The three peaks shown are the effective recombination cross sections, measured as a function of center-

Stimulated Recombination
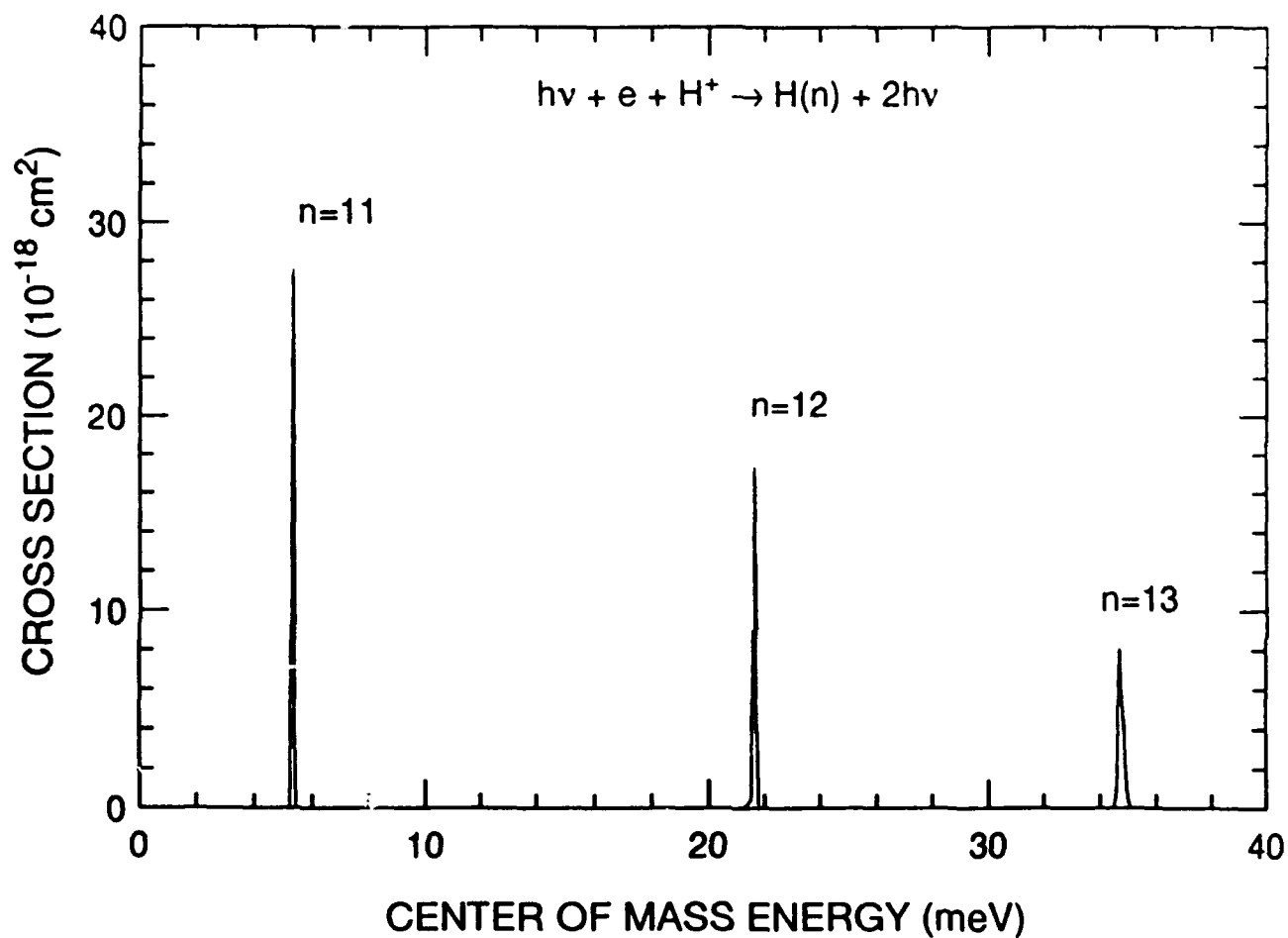
4.   Effective cross sections for the stimulated radiative recombination of electrons and protons to form hydrogen atoms in the states n=11, 12 and 13.

of-mass collision energy, normalized to 5 watts of laser power. Figures 5, 6, and 7 show these individual peaks using an expanded scale.

It can be seen that the energy resolution achieved in these experiments is very high; the peak widths being in the range from 0.3 meV to 0.5 meV. Table I lists the cross sections calculated using equation 1 with the corresponding measured cross sections for the stimulated process. Also shown are the gains calculated using equation 2. It can be seen that while the gains measured for n=12 and 13 are close to that expected, that for n=11 is low by a factor of about 3. It is not clear why this gain is low but it may be that the energy stability of the apparatus is not good enough to be able to sit on the exact peak for sufficient time to make the measurements. This effect is especially problematic at the low collision energies where the n=11 peak lies.

A study has been made of the relationship between the gain and the laser power over the range from 0 to 15 watts and the results are shown in figure 8. It can be seen that over this energy range the relationship is linear and there is no sign of re-ionization leading to a gain reduction.
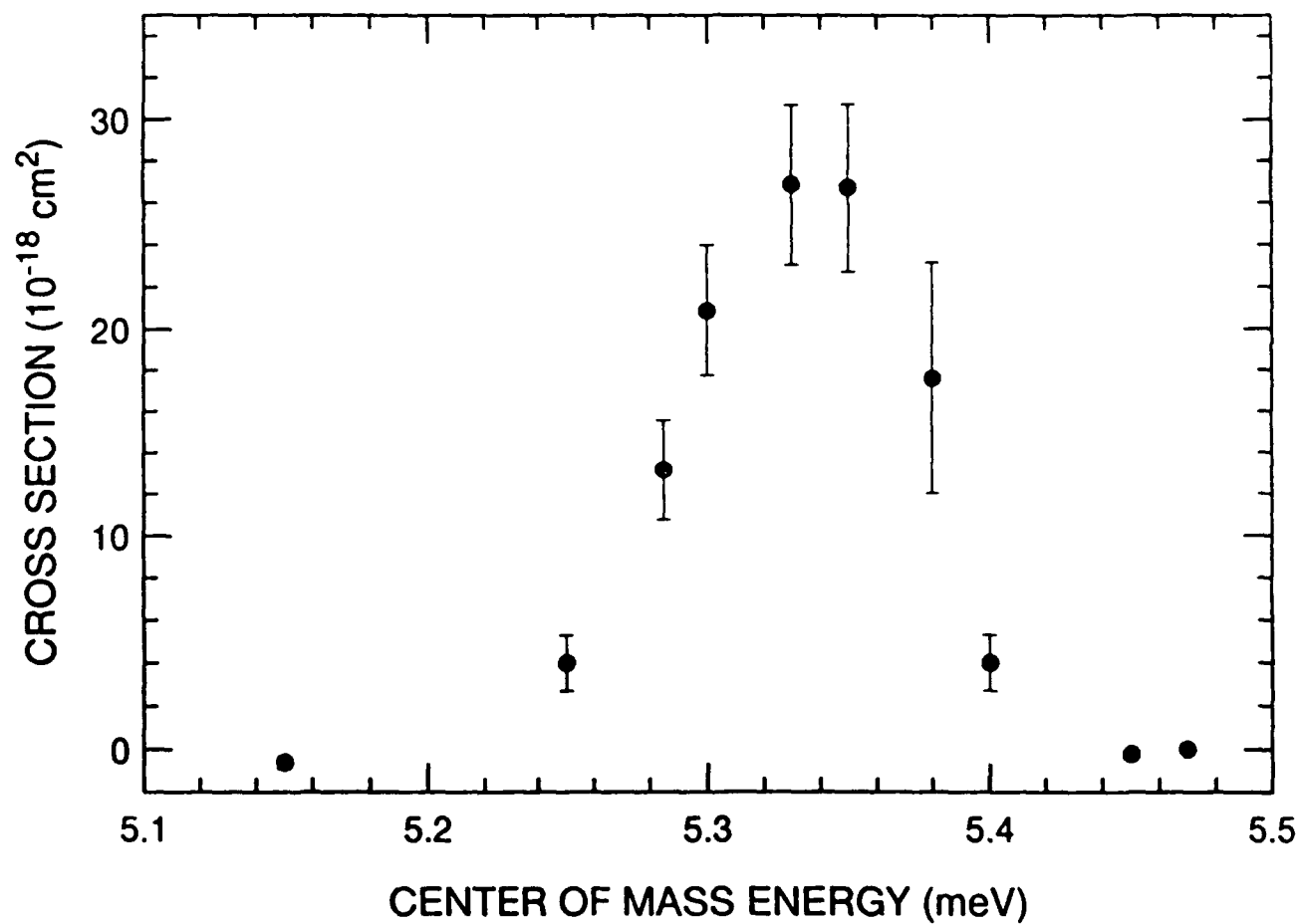
2. Electron-He⁺ Stimulated Recombination

Initial measurements have been made of the process:

$$hv + e + He^+ \rightarrow He(nl) + 2hv \ldots\ldots\ldots(III)$$

and the results of these measurements are shown in figures 9, 10 and 11 for capture into the n=15 and n=16 states. In this case we obtain double peaked resonances. This may be possibly explained as follows. The neutral helium spectrum consists of two series; a singlet and a triplet, figure 12. The triplets lie lower in energy than the singlets and so for
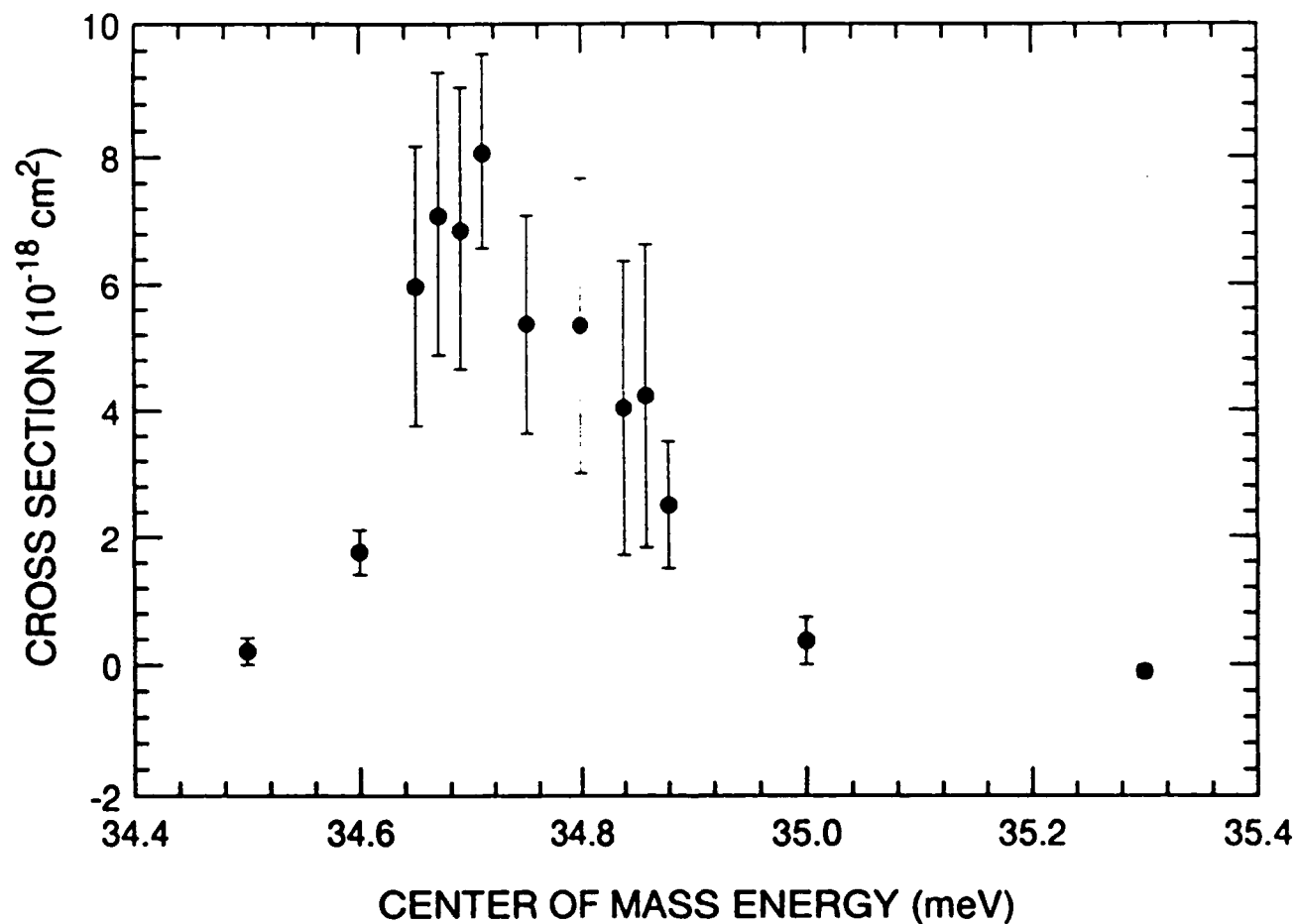
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N=11

5.   Effective cross sections for the stimulated recombination of electrons and protons to form hydrogen atoms in the n=11 state. (Expanded scale).

N = 12
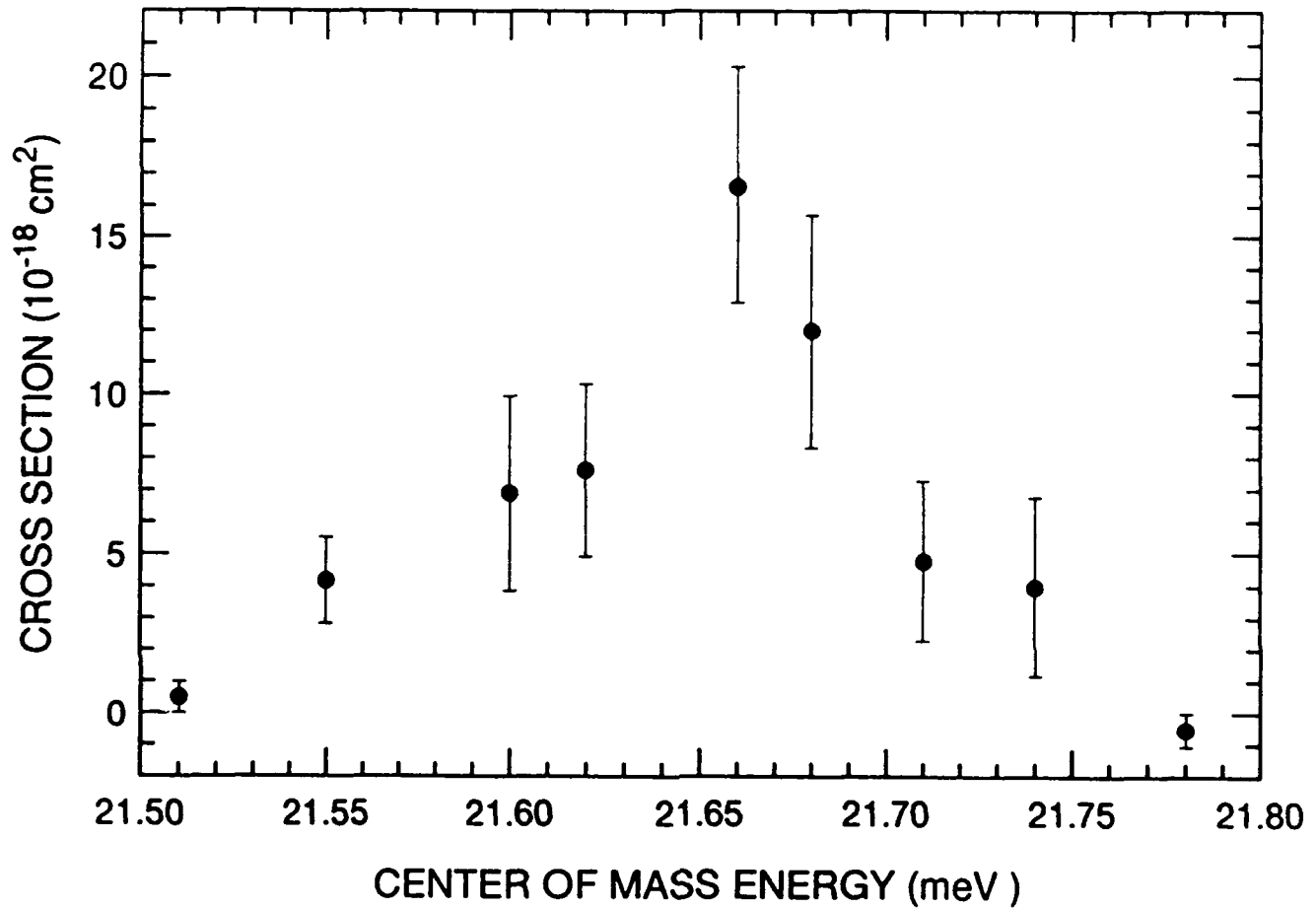
6.   Effective cross sections for the stimulated recombination of electrons and protons to form hydrogen atoms in the n=12 state. (Expanded scale).

N=13

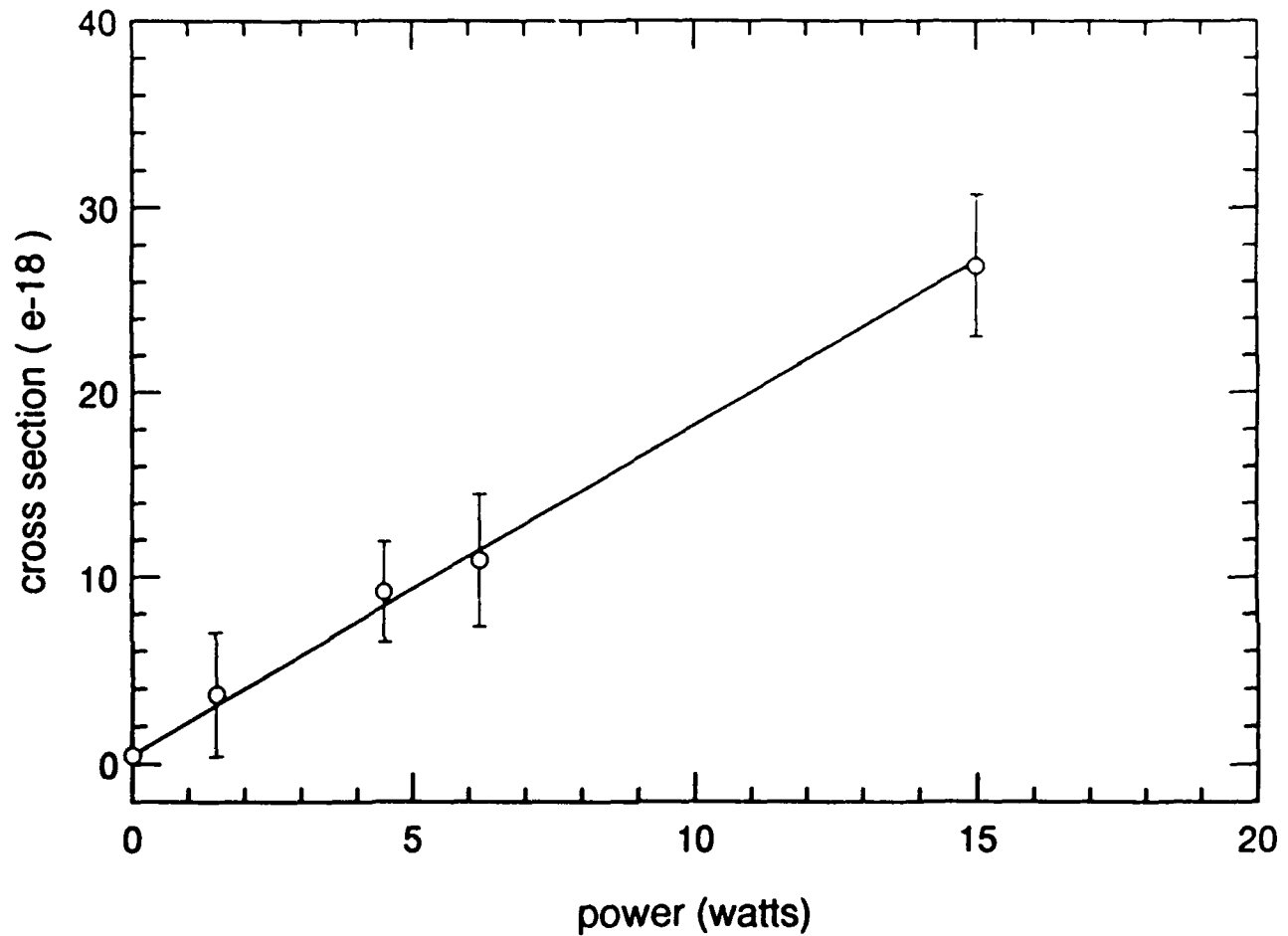7. Effective cross sections for the stimulated recombination of electrons and protons to form hydrogen atoms in the n=13 state. (Expanded scale).

Gain vs. Power (n=11)

8.   Measured relationship between gain and laser power for H^+ stimulated recombination.

a given photon energy, stimulated recombination to these states will occur at an energy less than to the singlets. These results are very new and there has not been time prior to the writing of this report to verify this explanation.

The uniqueness of the stimulated recombination process and the high energy resolution of the merged beam technique combine beautifully in this example to demonstrate the fine details of atomic spectroscopy.

3. Laser Induced Effects in Dissociative Recombination

For most small molecular ions, the dissociative recombination process is believed to proceed either directly via the formation of a repulsive intermediate state:

$$AB^+ + e \rightarrow AB^{**} \rightarrow A^* + B \ ...(I)$$

or indirectly via the formation of a vibrationally or rotationally excited Rydberg state that is subsequently predissociated:

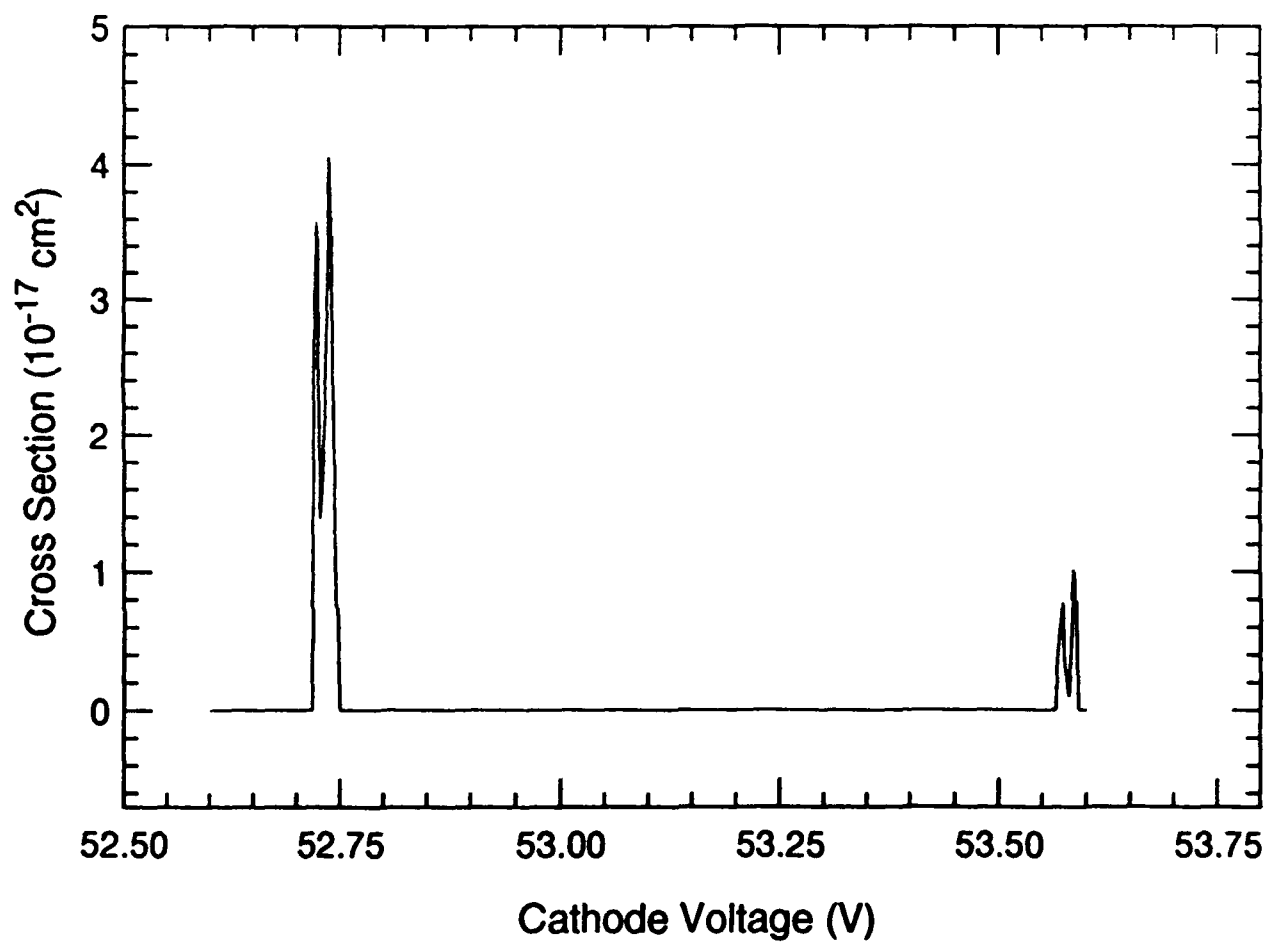$$AB^+ + e \rightarrow AB^R \rightarrow AB^* \rightarrow A^* + B \ ...(II)$$

For H_2^+, the repulsive state intersects the ion ground state in the vicinity of the v=1 level and mechanism (I) dominates over mechanism (II). For H_3^+ on the other hand, there is a non-optimal crossing between the ion ground state and the repulsive state and so mechanism (II) must dominate. Previous studies[5] performed using the merged beam technique have indicated that some of the intermediate states formed during the recombination of H_3^+ have lifetimes of 10^{-7} s or greater. Since these states are highly excited, it was assumed that it should be possible to perturb them via photon absorption and thus possibly to alter the cross section for the recombination process. An experiment

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TABLE I. SUMMARY OF EXPERIMENTAL RESULTS FOR STIMULATED RADIATIVE RECOMBINATION OF H^+
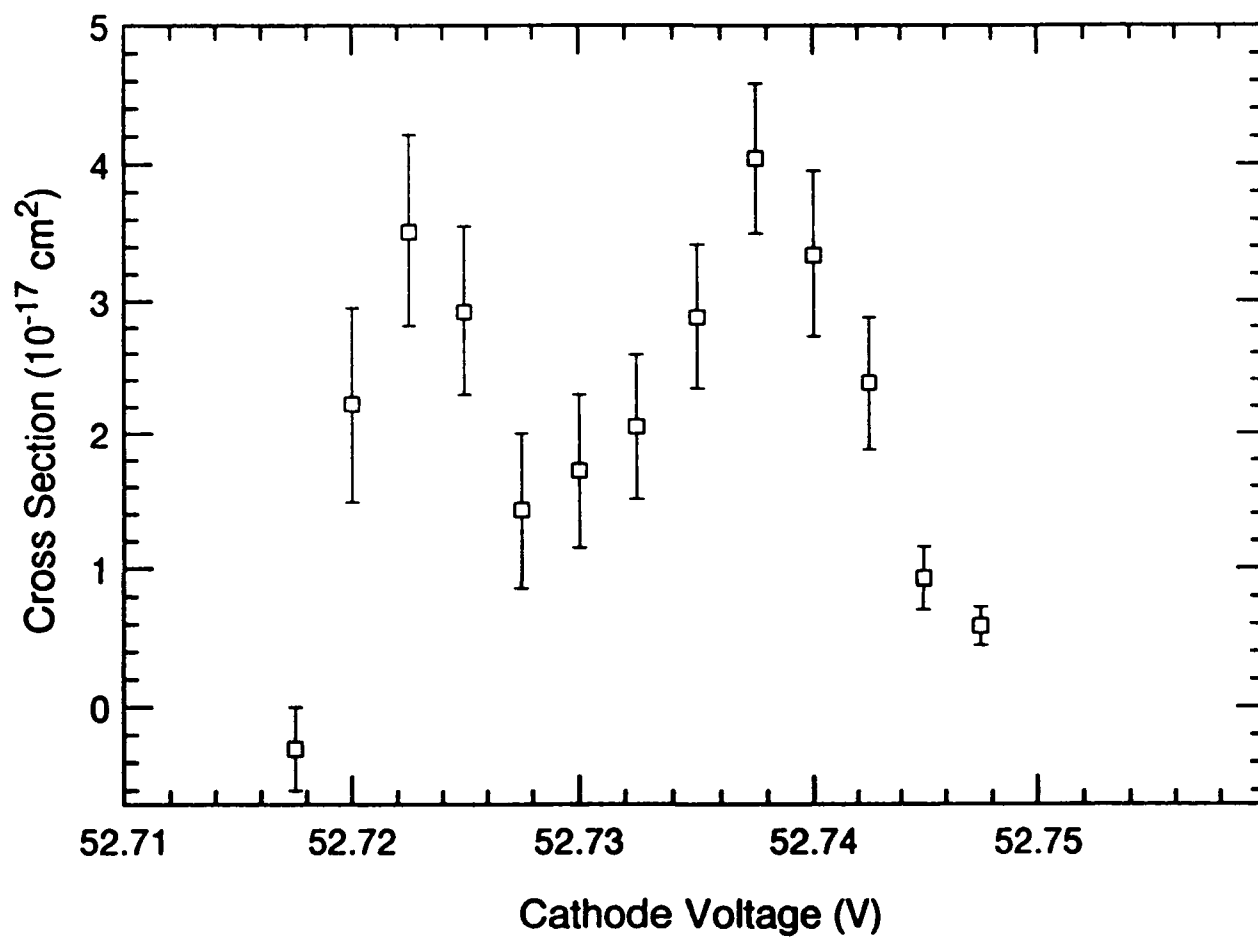
| n	σ_{calc}	σ_{meas}	Gain $\sigma_{meas}/\sigma_{calc}$	ΔE_{cm} (meV)	Gain$_{theor}$
11	4.6×10^{-20}	2.8×10^{-17}	603	0.3	2300
12	8.9×10^{-20}	1.7×10^{-17}	1910	0.3	2300
13	4.4×10^{-21}	8.0×10^{-18}	1810	0.5	1400

He Stimulated Recombination

9.   Effective cross sections for the stimulated recombination of electrons and He[+] ions to form helium atoms in the n=15 and n=16 states.

He Stimulated Recombination

10.   Effective cross sections for the stimulated recombination of electrons and He$^+$ ions to form helium atoms in the n=15 state.
(Expanded scale).

He Stimulated Recombination
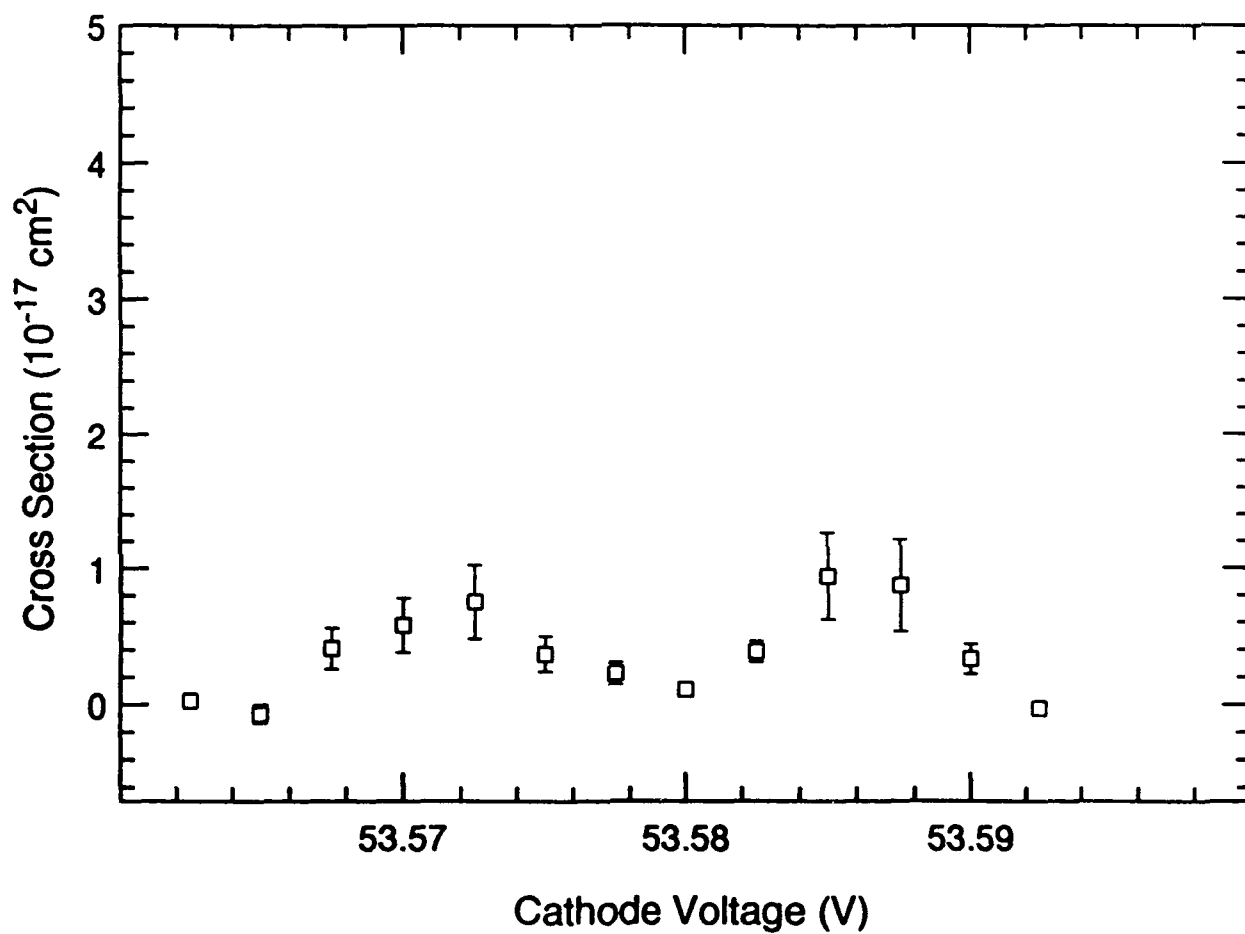
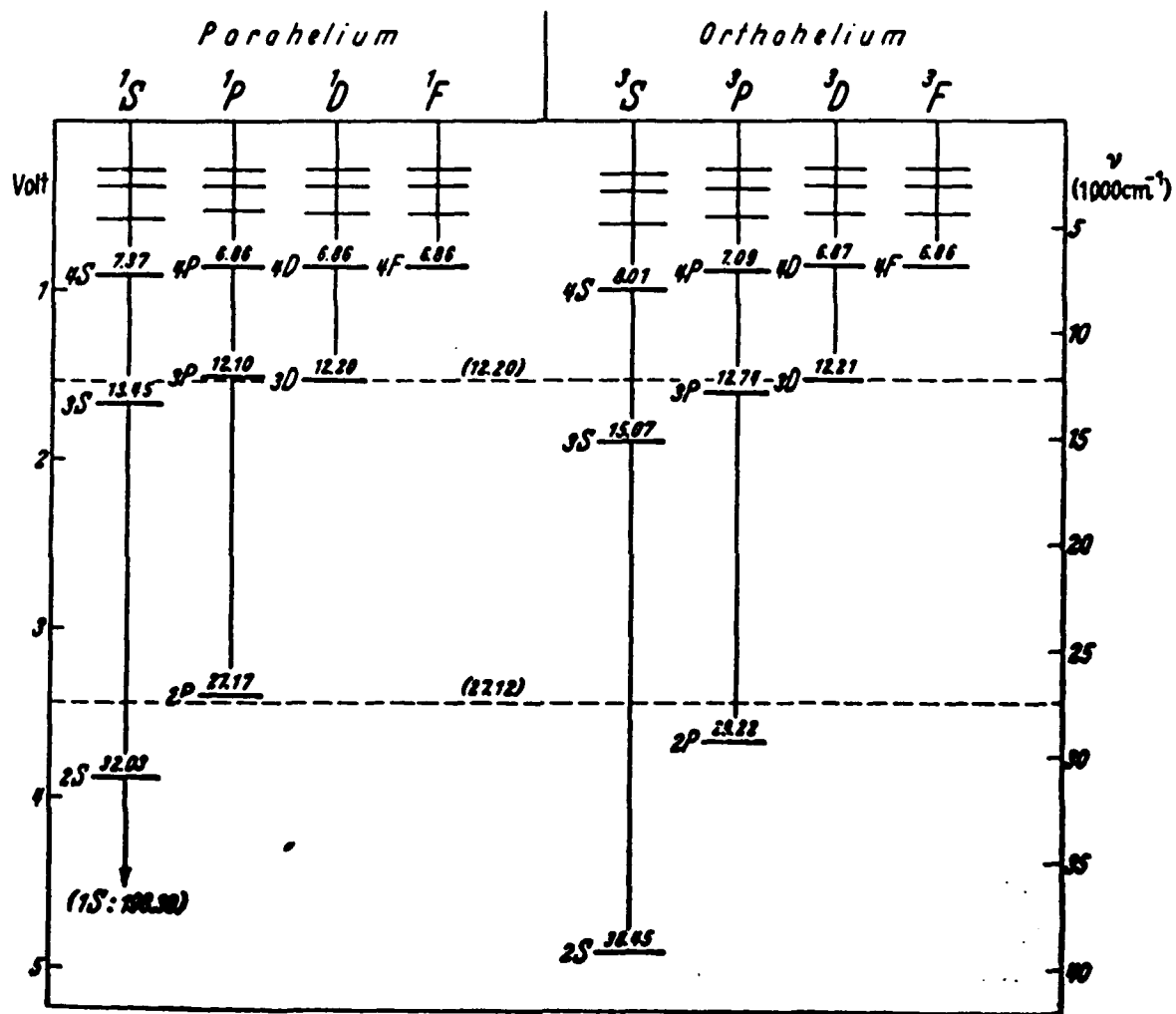11.   Effective cross sections for the stimulated recombination of electrons and He⁺ ions to form helium atoms in the n=16 state.
(Expanded scale).

Parahelium | Orthohelium

^1S ^1P ^1D ^1F ^3S ^3P ^3D ^3F

Volt

ν (1000cm⁻¹)

4S 7.37   4P 6.86   4D 6.86   4F 6.86   4S 8.01   4P 7.09   4D 6.87   4F 6.86

1

5

10

3S 13.45   3P 12.10   3D 12.20   (12.20)   3P 12.74   3D 12.21

15.07

2

15

3

25

2P 22.17   (22.12)   2P 23.22

2S 32.03

4

30

(1S :)

35

2S 38.45

5

40

12.   Energy level diagram for neutral HeI showing the singlet and triplet manifolds.

Laser Interrupted DR
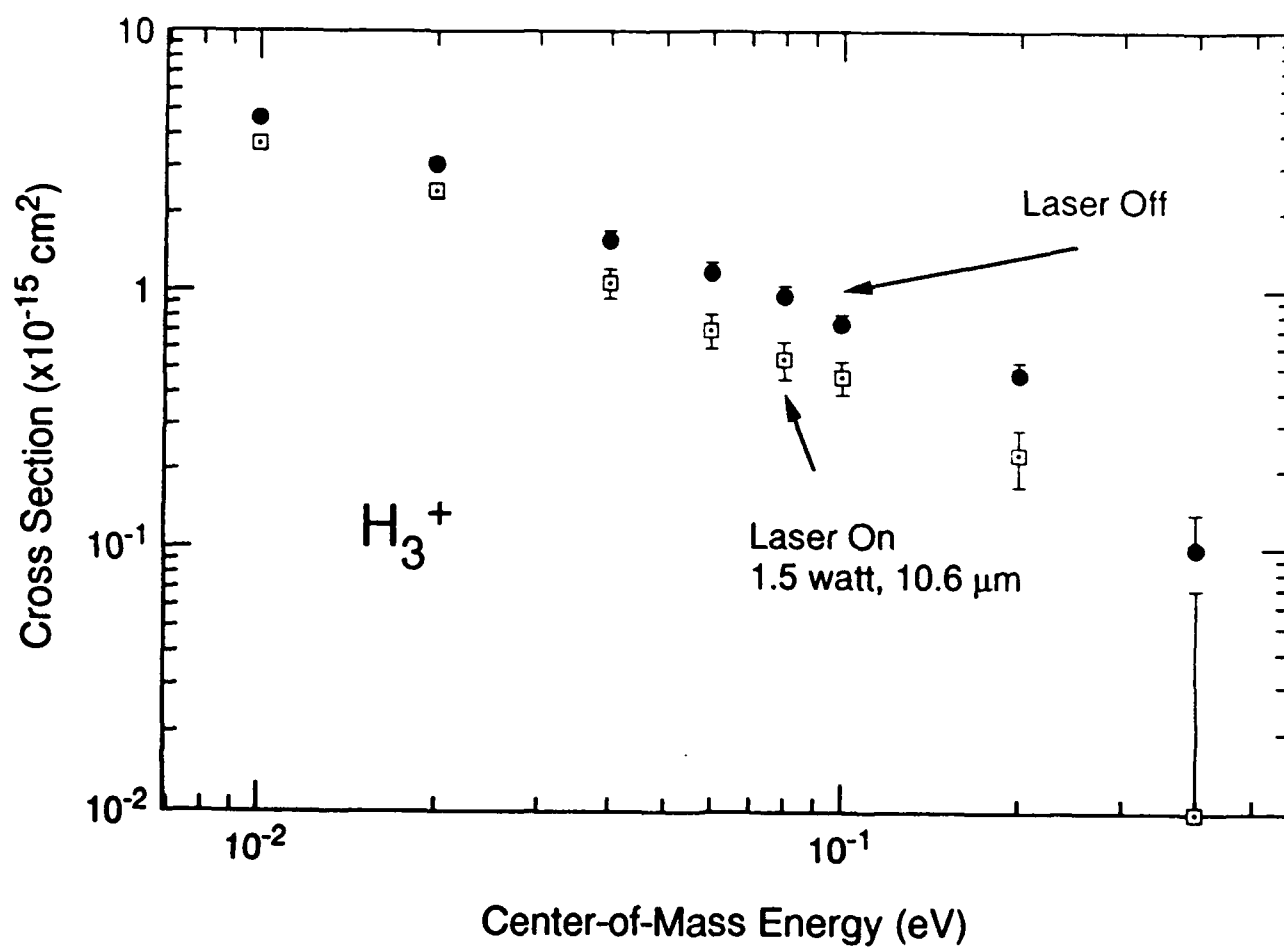
13. Cross section for the recombination of H_3^+ ions with and without laser irradiation.

Laser Interrupted DR
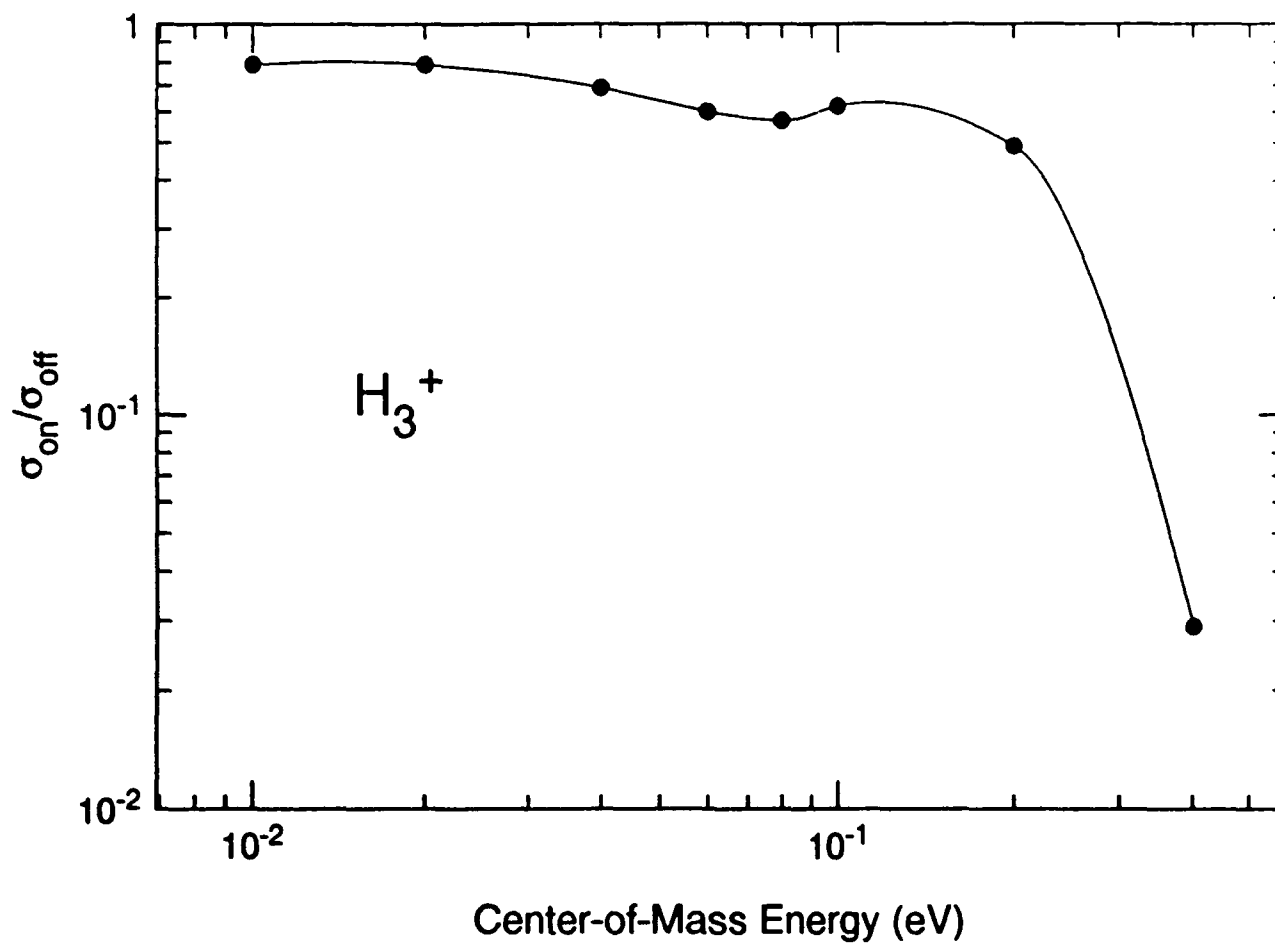
14.   Ratio of σ_{on}/σ_{off} for the dissociative recombination of H_3^+ with and without laser irradiation.

has been performed using the techniques and apparatus described above, to investigate this possibility.

The results are shown in figures 13 and 14. When 5 watts of power are used, the cross section at low energies is reduced by a factor of 0.8 while above 0.4 eV, the cross section goes to zero. Surprisingly however, when the laser power is increased above 5 watts, the cross section returns to its original value and does not change with further increases in power. The reason for this is not at all clear at this time and further studies of this peculiar phenomenon are planned.

REFERENCES

1.   Bethe, H. and Salpeter, E.E. *Quantum Mechanics of One- and Two- Electron Atoms* (Plenum, New York), 1977.

2.   Neumann, R., Poth, H., Winnacker, A. and Wolf, A. Z. Phys. A. **A313**, 253, 1983.

3.   Yousif, F.B., Van der Donk, P., Kucherovsky, Z., Reis, J., Brannen, E., and Mitchell, J.B.A. Phys. Rev. Lett. **67**, 26, 1991.

4.   Schramm, U., et al. Phys. Rev. Lett. **67**, 22, 1991.

5.   Mitchell, J.B.A. and Yousif, F.B. in *Microwave and Particle Beam Sources and Directed Energy Concepts* (ed. H.E. Brandt) Proceedings of SPIE Vol. 1061, SPIE, Bellingham, Washington, 1989, p. 536.